147 USPQ 391

In re WESSLAU

U.S. Court of Customs and Patent Appeals

Appl. No. 7447

Decided November 26, 1965

353 F2d 238

Headnotes

PATENTS

[1] Patentability--Composition of matter (► 51.30)

Claims to process of polymerizing ethylene are not rejected on theory that applicant's catalyst system can be met merely by substitution of groups from two prior patents on the corresponding components of a third prior system since no one of the references suggests such a substitution, quite apart from the result which would be obtained thereby; such piecemeal reconstruction of prior art patents in light of applicant's disclosure is contrary to 35 U.S.C. 103.

[2] Patentability--Invention--In general (► 51.501)

Question in cases within ambit of 35 U.S.C. 103 is whether subject matter as a whole would have been obvious to one of ordinary skill in the art following teachings of prior art at time invention was made; it is impermissible within framework of section 103 to choose from any one reference only so much of it as will support a given position, to exclusion of other parts necessary to full appreciation of what reference fairly suggests to one of ordinary skill in the art.

Particular Patents

Particular patents--Polyethylene

Wesslau, Process for the Production of Polyethylene with Narrow Distribution of the Molecular Weight, claims 35 to 43 of application allowed.

Case History and Disposition

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Appeal from Board of Appeals of the Patent Office.

Application for patent of Hermann Wesslau, Serial No. 753,872, filed Aug. 8, 1959; Patent Office Group 140. From decision rejecting claims 35 to 43, applicant appeals. Reversed.

Attorneys

ARNOLD SPRUNG, New York, N.Y., and ARNOLD B. CHRISTEN, Washington, D. C., for appellant.

CLARENCE W. MOORE (FRED W. SHERLING of counsel) for Commissioner of Patents.

Judge

Before WORLEY, Chief Judge, and RICH, MARTIN, SMITH, and ALMOND, Associate Judges.

Opinion Text

Opinion By:

ALMOND, Judge.

This appeal is from the decision of the Board of Appeals affirming the rejection of claims 35-43 [1] in appellant's application [2] entitled "Process for the Production of Polyethylene With Narnow Distribution of the Molecular Weight." No claims have been allowed.

[1] Appellant withdrew the appeal with respect to the only product claim 44, which was drawn to a polyethylene having a narrow molecular weight distribution characterized by a nonuniformity value U of magnitude between 2 and 4.

[2] Serial No. 753,872, filed August 8, 1958.

The invention relates to a process of polymerizing ethylene utilizing a Ziegler-type catalyst system to produce solid polyethylene. Both appellant and the Patent Office have treated the appealed process claims as standing or falling together, and we will do the same. Claim 35, from which the remaining claims depend, is illustrative and reads as follows:

> 35. In the process of polymerizing ethylene to a solid polymer having a high molecular weight and a narrow molecular weight distribution range, the improvement which comprises polymerizing ethylene in the presence of a polymerization catalyst con
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> sisting essentially of a mixture of titanium trichloride, at least one compound of tetravalent titanium $Ti(R)_4$ and at least one organic aluminum compound soluble in a liquid hydrocarbon and having the general formula $R'Al(R)_2$ in which R' is alkyl and R is selected from the group consisting of halogen, alkoxy and aroxy radicals, wherein between said tetravalent titanium compound and said organic aluminum compound there is present in said mixture at least one halogen atom and at least one member selected from the group consisting of alkoxy and aroxy radicals.

According to appellant's disclosure, polyethylene of high molecular weight may be produced by what has become known in the art as the Ziegler polymerization process. Analysis of the polyethylene so produced has revealed that although the *average* molecular weight of the polymer is high, a fairly large proportion of the individual polymer chains have a relatively low molecular weight. These low molecular weight fractions are particularly unfavorable for such properties as impact bending strength, rubbing, and fatigue. Appellant has discovered that the proportion of the lower molecular weight chains can be reduced, thereby narrowing the molecular weight distribution, by employing a three-component catalyst system in which either the $Ti(R)_4$ or $R'Al(R)_2$ contains an alkoxide or aroxide moiety.

The references relied on are:

Anderson 2,862,917 December 2, 1958

Muehlbauer 2,905,661 September 22, 1959

Ruhrchemie (Belgian) 553,694 June 24, 1957

The Ruhrchemie patent relates to a process for producing polyethylene of a desired molecular weight employing certain specified catalyst systems. The pertinent portion of the patent specification reads as follows:

> * * * when high molecular weight [polyethylene] products are to be obtained * * *, the employed mixtures consist of aluminum alkyl compounds and/or halides of aluminum alkyl with quantities of titanium trichloride of at least 0.01 mole * * * and quantities of titanium tetrachloride lower than 0.01 mole * * *; on the other hand, when materials having low molecular weight are to be obtained the employed mixtures consist of aluminum alkyl and/or halide of aluminum alkyl with more than 0.1 mole * * * of titanium tetrachloride per mole of aluminum alkyl and/or halide of aluminum alkyl, and with titanium trichloride at the rate of at least 0.1 mole, preferably 0.3-1 mole approximately per mole of aluminum alkyl and/or halide of aluminum alkyl.

The Anderson patent relates to a process of polymerizing ethylene whereby control over the weight average molecular weight of the polymer and the *molecular weight distribution* of the polymer is achieved by adhering to process conditions which insure the solubility of the ethylene during polymerization. The process employs coordination catalysts of titanium:

> * * * obtained by admixing a trivalent or tetravalent titanium compound of the class consisting of titanium salts and titanium alkoxides with a compound having at least one metal-to-hydrocarbon bond, such as metal alkyls, suitable compounds being lithium aluminum alkyls, aluminum alkyls, Grignard reagents, alkyl aluminum halides, tin alkyls, etc. * * *

Anderson further states:

> * * * the steady state compliance [an indicia of molecular weight distribution] will vary from 3 to 7 when the critical conditions of the process of the present invention are maintained and will rise to a range of 12 to 28 when the polymerization is carried out at conditions other than required by the process of the present invention. * * *

Muehlbauer relates to a process for producing high molecular weight polyolefins employing a two-component catalyst system consisting of certain metal halides and a compound of the formula XAlR(OR'), where X is halogen, and R and R' are the same or different alkyl, cycloalkyl, or aryl radicals. Titanium trichloride and titanium tetrachloride are specifically disclosed as suitable metal halides.

The sole issue in this case is obviousness under 35 U.S.C. 103.

Appellant's principal contention is that:

> * * * since none of the reference[s] either singly or in combination teach a control of the molecular weight distribution range by specific selection of catalyst components, or even that the nature or composition of the catalyst could have an effect on this molecular weight distribution range, the subject matter of the invention as a whole could not possibly be obvious from the references. * * *

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We agree. Appellant's specification contains ten examples in which various three-component catalyst systems were utilized in the polymerization of ethylene. The systems set forth in three of these examples consisted of (1) titanium trichloride, (2) titanium tetrachloride, and (3) diethyl aluminum monochloride is various molar ratios. These fall within the catalyst systems disclosed by Ruhrchemie. The U value, which according to appellant's specification is a measure of the molecular weight distribution, ranges from 6.3 to 12.8 for such catalysts. In the remaining seven examples, catalyst systems covered only by the appealed claims were employed, with the nonuniformity value U [3] for the resultant polyethylene ranging from 2.6 to 3.9 We believe this to be a convincing demonstration that the alkoxide or aroxide moiety, when present in the catalyst systems of the appealed claims, possesses the property of conferring a significant degree of control over the ultimate molecular weight distribution of polyethylene. This property is neither taught nor suggested by the prior art.

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[3] Appellant's specification contains the following description of the nonuniformity value U:

> * * * the so-called non-uniformity is used for characterising the range of distribution of the molecular weights. According to G. V. Schulz in H. A. Stuart's Die Physik der Hochpolymeren, 2nd vol., the macromolecule in solutions is given on page 754 as:

Graphic material consisting of a complex mathematical formula set at this point is not available. See text in hard copy or call BNA at 1-800-372-1033.

'M_w and 'M_n can be calculated from the molecular weight distribution by current methods (G. V. Schulz and M. Marx: Makromolekulare Chemie XIV (1954), pages 53-64).

The reasoning of the examiner and the board appears to be as follows: Ruhrchemie discloses a titanium trichloride - titanium tetrachloride - mono - ethyl aluminum dichloride system. This differs from appellant's system only in the latter's use of an alkoxide or aroxide group on either the tetravalent titanium or aluminum component or both. Since Anderson shows a tetravalent titanium compound containing an alkoxide group and Muehlbauer shows an aluminum compound containing an alkoxide group, appellant's catalyst system can be met merely by substitution of such alkoxide groups on the corresponding components of the Ruhrchemie system.

[1] The fallacy of this reasoning is that no one of the references *suggests* such a substitution, quite apart from the result which would be obtained thereby. Such piecemeal reconstruction of the prior art patents in the light of appellant's disclosure is contrary to the requirements of 35 U.S.C. 103. In re Rothermel, 47 CCPA 866, 276 F.2d 393, 125 USPQ 328 .

[2] The ever present question in cases within the ambit of 35 U.S.C. 103 is whether the subject matter as a whole would have been obvious to one of ordinary skill in the art following the *teachings* of the prior art at the time the invention was made. It is impermissible within the framework of section 103 to pick and choose from any one reference only so much of it as will support a given position, to the exclusion of other parts necessary to the full appreciation of what such reference fairly suggests to one of ordinary skill in the art. The Anderson patent is the only reference before us which recognizes the desirability of producing polyethylene with a narrow molecular weight distribution range. Were one to follow the teachings of that patent in its entirety, he would be led to believe that control over the molecular weight distribution of polyethylene was gained independently of the catalyst system, a belief untenable in light of

appellant's disclosure.

Both the board and the solicitor apparently assert the position that it is incumbent upon appellant to show that his results are outstanding as compared with the results accomplished by Anderson and Muehlbauer. If this is construed as requiring appellant to show unexpected results accruing from his claimed process, we think he has met the requirement. We perceive no teaching in the prior art of record suggesting that an alkoxide or aroxide moiety in a Ziegler-type catalytic system would produce the results obtained by appellant's process.

The decision of the board is reversed.

- End of Case -

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